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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
10/029,891	12/21/2001	Richard C. Willson III	2000-046CON2	5096

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SYMYX TECHNOLOGIES INC
LEGAL DEPARTMENT
3100 CENTRAL EXPRESS
SANTA CLARA, CA 95051

EXAMINER	
SODERQUIST, ARLEN	

ART UNIT	PAPER NUMBER
1743	

6

DATE MAILED: 02/10/2004

Please find below and/or attached an Office communication concerning this application or proceeding.

Office Action Summary	Application No. 10/029,891	Applicant(s) WILLSON, RICHARD C.	
	Examiner Arlen Soderquist	Art Unit 1743	

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

Period for Reply

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE _3_ MONTH(S) FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If the period for reply specified above is less than thirty (30) days, a reply within the statutory minimum of thirty (30) days will be considered timely.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

Status

1)☒ Responsive to communication(s) filed on _19 November 2003_.
2a)☐ This action is **FINAL**. 2b)☒ This action is non-final.
3)☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

Disposition of Claims

4)☒ Claim(s) _1-31_ is/are pending in the application.
4a) Of the above claim(s) _____ is/are withdrawn from consideration.
5)☐ Claim(s) _____ is/are allowed.
6)☒ Claim(s) _1-31_ is/are rejected.
7)☐ Claim(s) _____ is/are objected to.
8)☐ Claim(s) _____ are subject to restriction and/or election requirement.

Application Papers

9)☐ The specification is objected to by the Examiner.
10)☐ The drawing(s) filed on _____ is/are: a)☐ accepted or b)☐ objected to by the Examiner.
Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).
Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).
11)☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

Priority under 35 U.S.C. § 119

12)☐ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).
a)☐ All b)☐ Some * c)☐ None of:
1.☐ Certified copies of the priority documents have been received.
2.☐ Certified copies of the priority documents have been received in Application No. _____.
3.☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).
* See the attached detailed Office action for a list of the certified copies not received.

Attachment(s)

1) ☒ Notice of References Cited (PTO-892)
2) ☐ Notice of Draftsperson's Patent Drawing Review (PTO-948)
3) ☐ Information Disclosure Statement(s) (PTO-1449 or PTO/SB/08) Paper No(s)/Mail Date _____.
4) ☐ Interview Summary (PTO-413) Paper No(s)/Mail Date. _____ .
5) ☐ Notice of Informal Patent Application (PTO-152)
6) ☐ Other: _____.

1. Applicant is advised that should claim 7 be found allowable, claim 26 will be objected to under 37 CFR 1.75 as being a substantial duplicate thereof. When two claims in an application are duplicates or else are so close in content that they both cover the same thing, despite a slight difference in wording, it is proper after allowing one claim to object to the other as being a substantial duplicate of the allowed claim. See MPEP § 706.03(k).

2. The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

> (a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negatived by the manner in which the invention was made.

The factual inquiries set forth in *Graham* v. *John Deere Co.*, 383 U.S. 1, 148 USPQ 459 (1966), that are applied for establishing a background for determining obviousness under 35 U.S.C. 103(a) are summarized as follows:

1. Determining the scope and contents of the prior art.
2. Ascertaining the differences between the prior art and the claims at issue.
3. Resolving the level of ordinary skill in the pertinent art.
4. Considering objective evidence present in the application indicating obviousness or nonobviousness.

3. Claims 1, 4-13, 18-20, 22-31 and 2 as it depends from claim 30 are rejected under 35 U.S.C. 103(a) as being unpatentable over Carlson in view of Gimezewski (newly cited and applied), Johnson (Analytical Calorimetry, newly applied), Kulkova, Schödel (DD 234942 translation attached to this office action) or Temkin. In the paper Carlson presents apparatus and a microactivity test for measuring low temperature activity of automobile exhaust catalysts. In the first paragraph of page 467, a need is taught for rapid screening tests and in particular "tests that will measure low temperature activity" to reduce the amount of costly testing required for catalyst research and development. In the second paragraph of the same page, differential thermal analysis (DTA) was taught as a suitable tool for measuring low temperature catalytic activity. Added benefits include the ability to rate a catalyst directly against other catalysts and the use of small samples. In the fourth paragraph of page 467, the aim of the research presented in the paper is taught as developing a rapid sensitive test giving data that can be interpreted relative to the performance of known and evaluated catalysts. A micro-activity test is described

in which differential thermal analysis is used to evaluate rapidly the ability of an auto exhaust catalyst to initiate and sustain combustion of a synthetic exhaust mixture. The study shows the flexibility of the test in that it provides direct comparison of experimental catalysts with inert standards, standard catalysts, or other experimental catalysts. Data are presented that demonstrate the capabilities of the test, the limitations of the test, and the hazards of over-interpretation of test results. Figure 1 shows the DTA microactivity flow apparatus used including thermocouples. In the last paragraph of page 468, the criteria of low temperature activity used to compare or rate the catalysts is given. The criteria used was the temperature that the exotherm appears and the rate of temperature rise to the exotherm maximum. The use of the temperature that the exotherm maximum is observed should be used in conjunction with the initiating or "light-off" temperatures since both low "light-off" temperatures and rapid heat evolution or rate of conversion are necessary for good low temperature catalytic activity. The test reproducibility was demonstrated and found to be more than adequate for catalyst screening purposes. Table 3 with its associated discussion and figures explains how the rating or comparison would happen. Figures 10-11 and their related discussion teach a possible thermal stability test. Figure 9 and its discussion show the use of a known catalyst as a reference in the method and device. Carlson does not teach simultaneous measurement of a plurality of catalyst candidates at a plurality of sites on a common support.

In the paper, Gimzewski teaches a multi-sample high-pressure DTA for measuring oxidation induction times. The design and operation are described of a DTA instrument which can run five samples simultaneously to measure their oxidation induction times in oxygen or air at pressures up to 70 bar and isothermal temperatures up to 250°. Although designed for high-productivity testing of automotive lubricants, the instrument is applicable to other materials such as greases, edible oils and polymers. The third to fifth paragraphs of page 98 discuss the multi-sample concept and its use in the described DTA device because of the considerable demand for DTA testing in the petroleum industry. The described device is a batch reactor (see page 101, "static atmosphere" under the "Gas flow arrangement" heading. The conclusion (page 105) teaches that the device has produced substantial improvements in accuracy and productivity and points to an expectation of similar improvements with other materials.

In the paper Johnson teaches multiple sample differential scanning calorimetry with

a new multiple sample differential scanning calorimeter (DSC) system, incorporating a microprocessor-based data system. It can perform measurements on three samples in a single furnace simultaneously in the time usually required to do one. Page 133 lists four limitations with known DTA apparatus that are overcome in the described apparatus. Because all samples experience the same thermal history, more dependable intercomparisons of samples may be made by including reference or quality control materials and calibration standards. Also see page 139 for a listing of six advantages of the multi-sample system. The data system provides teletype control of all aspects of the experiment except the furnace program; it stores the data, makes identification labels, and draws the DSC curves. The system comprises a multiple sample, single reference DSC cell, a data system, a furnace-controller and a recorder as shown in figures 1-2 and 5. The design and performance of the system are described.

In the paper Kulkova presents apparatus for testing catalysts for the oxidation of ethylene to ethylene oxide. A tubular reactor for the simultaneous testing of ≤ 5 catalyst samples (5 cc. each) under isothermal conditions is described. In the oxidation of C_2H_4 at 220-320°, a Ag catalyst with Se promoter was used for ≤ 2800 hours with good results, the catalyst productivity being between 115 and 330 g./l. hour. The reaction products were analyzed by gas chromatography in a 200-mm. column, with polyethylene glycol sebacate supported on Celite-545. Pages 2-4 of the translation describe the apparatus including the presence of thermocouples (paragraph bridging pages 2-3).

In the published application Schödel teaches method and process for determination of catalyst activity. Catalyst activity is routinely, rapidly, and accurately monitored in a sampling-testing system in which 3-5 samples, containing 0.05-2 g catalyst each, are heated separately and simultaneously in a radiation oven, separate 2-30 L/h gas flows (containing reactants and withdrawing products) are passed through the samples, and the products are withdrawn and separately injected for analysis; each sample has a separate temperature-measuring element and the temperature of each sample can be independently controlled and monitored by a rotating dial. Thus, 3 samples (containing 200 mg each) of a Pt/Al_2O_3 alkane reforming catalyst were reduced in hydrogen and tested for reforming of heptane (at 1.15 g/g catalyst-h flow); the 3 samples had heptane conversions and yields of aromatic

hydrocarbons of 46.4 ± 0.7 and 22.9 ± 0.4, 28.7 ± 0.9 and 12.4 ± 0.2, and 36.9 ±0.8 and 23.5 ± 0.4 mol%.

In the abstract Temkin presents an ideal-displacement laboratory reactor. A laboratory reactor is described having a set of small diameter tubes. Catalyst granules or pellets are placed into the tubes, diluted with an inert material, and the catalytic activity for a heterogeneous reaction can be investigated simultaneously for several catalysts, depending on the number of tubes present. The wall of the tubes are equipped with thermocouples. Isothermal conditions are obtained by passing the gas through the system; the catalyst acquires the temperature of the tube walls.

It would have been obvious to one of ordinary skill in the art at the time the invention was made to modify the Carlson device to be able to run multiple samples simultaneously as taught by Gimezewski, Johnson, Kulkova, Schödel or Temkin because of the advantages of multi-sample processing as taught by Gimezewski, Johnson, Kulkova, Schödel and Temkin.

4. The nonstatutory double patenting rejection is based on a judicially created doctrine grounded in public policy (a policy reflected in the statute) so as to prevent the unjustified or improper timewise extension of the "right to exclude" granted by a patent and to prevent possible harassment by multiple assignees. See *In re Goodman*, 11 F.3d 1046, 29 USPQ2d 2010 (Fed. Cir. 1993); *In re Longi*, 759 F.2d 887, 225 USPQ 645 (Fed. Cir. 1985); *In re Van Ornum*, 686 F.2d 937, 214 USPQ 761 (CCPA 1982); *In re Vogel*, 422 F.2d 438, 164 USPQ 619 (CCPA 1970);and, *In re Thorington,* 418 F.2d 528, 163 USPQ 644 (CCPA 1969).

A timely filed terminal disclaimer in compliance with 37 CFR 1.321(c) may be used to overcome an actual or provisional rejection based on a nonstatutory double patenting ground provided the conflicting application or patent is shown to be commonly owned with this application. See 37 CFR 1.130(b).

Effective January 1, 1994, a registered attorney or agent of record may sign a terminal disclaimer. A terminal disclaimer signed by the assignee must fully comply with 37 CFR 3.73(b).

5. Claims 1-31 are rejected under the judicially created doctrine of obviousness-type double patenting as being unpatentable over claims 1-41 of U.S. Patent No. 6,063,633. Although the conflicting claims are not identical, they are not patentably distinct from each other because the instant claims are of a scope which completely encompasses the patented claims. For this reason one cannot practice the patented claims without infringing the claims in the instant application and the inventions are not distinct. The instant invention claims temperature sensors, but column

3 lines 1-8 and column 4 lines 45-55 of the patent make it clear that the infrared camera of the patented claims is a temperature sensor.

6. Applicant's arguments with respect to the claims have been considered but are moot in view of the new ground(s) of rejection.

7. The prior art made of record and not relied upon is considered pertinent to applicant's disclosure. The cited art relates to catalyst testing and multi-sample DTA instruments.

Any inquiry concerning this communication or earlier communications from the examiner should be directed to Arlen Soderquist whose current telephone number is (571) 272-1265 as a result of the examiner moving to the new USPTO location. The examiner's schedule is variable between the hours of about 5:30 AM to about 5:00 PM on Monday through Thursday and alternate Fridays.

A general phone number for the organization to which this application is assigned is (571) 272-1700. The fax phone number to file official papers for this application or proceeding is (703) 872-9306.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see http://pair-direct.uspto.gov. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).

Arlen Soderquist
February 4, 2004

ARLEN SODERQUIST
PRIMARY EXAMINER

PTO 03-3460

German Patent No. 234 942 A1

A METHOD AND ARRANGEMENT FOR DETERMINING CATALYST ACTIVITY

Rainer Schödel, et al.

UNITED STATES PATENT AND TRADEMARK OFFICE
WASHINGTON, D.C. MAY 2003
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A METHOD AND ARRANGEMENT FOR DETERMINING CATALYST ACTIVITY

[Verfahren und Anordnung zur Katalysatoraktivitätsbestimmung]

Inventors:	Dr. Rainer Schödel
	Michael Keck
	Ulrich Neumann
Patent Holder:	VEB Leuna Werke "Walter Ulbricht"
	Granted in accordance with §17 Paragraph 1 of the Patent Law

Claims

1. A method for determining catalyst activity in reactions with high heats of reaction with integral flow reactors in the temperature range from 293 to 993 K after prior catalyst activation, which is characterized by the fact that three to five parallel connected reaction tubes, which are heated in a radiant oven, are coated with 0.05 to 2 g of the various samples of catalysts, the gas flow rates are adjusted to 2 to 30 L/h in the reaction tubes with parallel connected throttles, the substrate to be reacted is fed to the central purified flow of gas leading to the flow regulators, sampling is simultaneously taken at the reaction tube outlets using a sampling device, the samples are supplied in succession to the gas chromatograph and temperature control of the

radiant oven takes place via thermoelements arranged in the catalyst bed, by sending the voltages of the thermoelements of the reaction tubes in succession to the set point controller for temperature control of the radiant oven by means of a rotating tap, where the transfer time for each thermoelement is 1 to 3 sec.

2. An arrangement for determination of catalyst activity, which is characterized by the fact that three to five reaction tubes are connected in parallel, a throttle is arranged in front of each reaction tube inlet, a substrate dispensing site with subsequent mixing vessel is connected to the central gas supply pipe leading to the parallel connected throttles, the reaction tube outlets are coupled to a multiple sampling device, the reaction tubes are arranged in a radiant oven and a thermoelement is arranged in each of the reaction tubes and the thermoelements are timewise linked via a rotating tap to the set point controller for temperature control of the radiant oven.

Application field of the invention

The invention concerns a method and arrangement for determining the catalyst activity of test catalysts and industrial catalysts for purposes of quality control and for preparation of catalysts in production plants for reactions with high heat of reaction.

Description of the known technical solutions

It has been known that the catalytic efficiency of catalysts is tested in laboratories and in industry by means of a microreactor technique. In many cases the microreactors are linked to gas chromatographs, with which analysis of the reaction products is undertaken. The use of such microreactor techniques has already been reported by E. Bayer (Angew. Chemie 69, 732 (1957)). This method is known from US Patent 2 905 539. In the catalytic tests the efficiency of the catalysts is tested in dependence on the temperature, pressure, gas composition and other test parameters. It is also known that for many heterogeneously catalyzed reactions multitube reactors are used in industry. In this case the reaction components arise in gas form at one end of the multitube reactor and flow through the individual tubes in which the catalyst is arranged. In doing so, the starting materials are converted and the gaseous reaction products exit at the reactor outlets and leave the reactor plant collected together.

BRD-OS [Offenlegungsschrift Patent Application], 2 714 939 utilizes an industrial multitube reactor to test the catalytic efficiency of catalysts, by first packing a tube with the test catalyst, collecting the gaseous reaction product leaving this tube, and then analyzing it.

This method can only conditionally be used to solve research problems and for quality control of industrial catalysts, since the cost of catalyst sampling is very high and precise control temperature is not possible. In addition, this method, like the traditional test devices, has the disadvantage that the expense of time, material and energy is relatively high.

Goal of invention

The goal of the invention is a method and an arrangement for determining the catalytic efficiency of catalyst in reactions that have high heats of reaction for purposes of quality control and for preparation of the catalyst batch in production plants with a low cost.

Nature of Invention

The invention is based on the task of developing a method and an arrangement for determining the activity of catalyst with heats of reaction that makes it possible to obtain reliable and expressive characteristic values for the catalytic activity of test and industrial catalysts with time, energy and material cost that is as low as possible.

This task is solved by a method for determining catalyst activity in reactions with high heats of reactions with integral flow reactors in the temperature range from 293 to 933 K after prior catalyst activation, by the fact that three to five parallel connected reaction tubes, which are heated in a radiant oven, are loaded with 0.05 to 2 g of the different catalyst samples, gas flow rates from 2 to 30 L/h are established in the reaction tubes with parallel connected throttles, the substrate to be reacted is dispensed into the central purified gas flow that leads to the flow regulators, and sampling is undertaken at the reaction tube outlets simultaneously using a sampling device. The samples are successively input to the gas chromatograph, and temperature control of the radiant oven takes place via thermoelements arranged in the catalyst bed, by sending the voltages of the thermoelements of the reaction tubes successively to the set point controller for temperature control of the radiant oven, where the transition time for each thermoelement is 1 to 3 sec. This task is further solved in accordance with the invention by an arrangement for catalyst activity determination in which three to five reaction tubes are arranged in parallel, a throttle is arranged in front of each reaction tube inlet, a substrate dispensing site with subsequent mixing baffle is connected to the center of gas supply line leading to the parallel connected throttles, and the reaction tube outlets are connected to a multiple sampling device. The reaction tubes are arranged in a radiant oven, a thermoelement is arranged in each of the reaction tubes, and the thermoelements are timewise connected to the set point controller for temperature control of the radiant oven via a rotating tap. Surprisingly, it was found that the brief successive temperature control of the individual reactors leads to temperature constancy such that reproducible catalytic characteristics of the catalyst can be obtained with the arrangement in accordance with the invention.

In addition, it is important for the successful application of the arrangement in accordance with the invention that the reaction tube outlets are connected to a sampling device, which carries out simultaneous sample taking and with which the samples can be successively sent to a gas chromatograph. It proved to be a good idea if the successive temperature measurement and thus

the temperature control in each reactor takes place over a time period of at least 2 sec. It is also important that a radiant oven be used as the oven. The principle of the arrangement can be used both for catalytic tests at normal pressure and for tests at elevated pressure. The gas adjustments and sample dispensing operations can be carried out manually or automatically. In the preferred embodiment of the arrangement in accordance with the invention the gas adjustments and dispensing operations take place automatically. With the initiation of the dispensing operation a microcomputer supported analysis value for determining the quantitative composition of the reaction products is set into operation. The invention is to be illustrated in more detail by means of an embodiment example.

Embodiment example

The three individual reactors of the multireactor unit are charged with the test catalysts A, B and C ($Pt-Al_2O_3$), with the initial catalyst weight being 200 mg. The catalysts are heated to 500°C in a radiant oven according to a temperature program, with feed of hydrogen, and held at this temperature for 1 h. Then an n-heptane/hydrogen mixture is passed through the catalyst bed. The catalyst load is 1.15 g n-heptane/g· h. The preparation of this mixture takes place by injecting n-heptane with a microdispensing device into the central hydrogen flow. A mixing device is connected inline with the injection site. The outlet of the mixing vessel leads to a gas distributor, to which three flow-control throttles are connected. The gas pressure in the pipes up to the flow regulated throttles is 0.2 MPa. The three reactors are connected to the flow controlled throttles via dips. Thermoelements project into the catalyst beds in the three reactors to measure and control the temperature there. The temperature control takes place so that the thermal voltages of the thermoelements of the individual reactors are fed to the set point controller for control of the radiant oven via a rotating tap, with the measurement time for each thermoelement being 2 sec. After reaction times of 30 and 90 min simultaneous sampling at the three reactor outlets is carried out using a multiple dispensing sampling device. The samples are automatically successively sent to the gas chromatograph for analysis of the reaction products.

Table 1 shows the results.

Table 1

Catalytic characteristics for the PT-Al_2O_3 catalysts characterized according to the embodiment example

Catalyst	n-heptane conversion in mol%	Formation of aromatics in mol%
A	46.4 ± 0.7	22.9 ± 0.4
B	28.7 ± 0.9	12.4 ± 0.2
C	36.9 ± 0.8	23.5 ± 0.4

The results show that reliable data are obtained in a short time and low cost with the test arrangement in accordance with the invention.